NATIONAL BUREAU OF STANDARDS REPORT

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INDOOR AIR POLLUTION STATUS REPORT

U.S. DEPARTMENT OF COMMERCE

NATIONAL BUREAU OF STANDARDS

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NBS PROJECT 4214101

April 19, 1971

NBS REPORT 10 591

INDOOR AIR POLLUTION STATUS REPORT

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1. Introduction

There are a number of questions relating to the quality of indoor air which have been indirectly raised by increased public concern over air pollution. For example, what protection, if any, do buildings offer to external air pollution? What pollutants are found indoors and what are their sources? Should pollution abatement filters be included in air conditioning systems? What is "fresh" air, and what is "stale" air? These and many other questions can be raised, but the answers require considerable guesswork, because indoor air pollution has not been extensively studied. While some progress has been made in the analytical methodology of air pollution, comparatively little attention has been devoted explicitly to indoor problems.

A project has been activated in the Environmental Engineering Section of the Building Research Division for the purpose of identifying indoor pollutants, both gaseous and particulates, comparing indoor/outdoor pollutant relationships, and developing experimental and mathematical methods for analyzing the way in which a building handles air pollutants. The purpose of this report is to describe the status of the work, to outline plans for future work, and to suggest areas where the analytical capability must be improved if a strong program in indoor air pollution is to be achieved.

2. Work Initiated in Study of Indoor Air Pollution

A. Literature Survey

A brief review of the literature on indoor air pollution prior to January, 1970, was prepared and is included as chapter 9 of the report prepared by the NBS Study Group on Air Pollution (1b). In addition to work dealing specifically with indoor air pollution there is extensive literature on analytical methodology which is pertinent to all aspects of air pollution. Some of the earlier methods

of analysis are reviewed in the annoted bibliography of Ruch (2) and the book by Jacobs (2). More recent work through 1968 is included in volume 2 of Stern's book (3). Chapter 6 of the NBS study group report also contains review material on chemical analytical aspects of air pollution (1a). Although there is room for improvement in specificity, sensitivity, and convenience of analytical methods, the state of the art is well enough developed to permit useful measurement of indoor pollution to be made.

B. "Black box" Model of a Building With Forced Ventilation

One approach which has been developed for the analysis of how a building handles external air pollution is the "black box" model. The model is applicable to a building with forced ventilation which operates under a slight positive pressure.

a. Derivation of Model

A rudimentary diagram of an enclosed space with forced ventilation which represents an air conditioned building is shown in figure 1. If Q is the total amount of pollutant in the enclosed space, and V is the total volume, the rate of change of Q with time is

$$\frac{dQ}{dt} = \underbrace{ab\,(1 - E)}_{\text{rate entering in fresh air}} + \underbrace{G}_{\text{rate generated in enclosed space}} - \underbrace{\frac{QEr}{V}}_{\text{rate removed by filtration}} - \underbrace{\frac{Qb}{V}}_{\text{rate removed in exhaust}} \qquad (1)$$

where a is the concentration of pollutant in outside air

b is the volume rate at which outside air is brought into the space

E is the efficiency of the filters

G is the rate at which pollutant is generated in the enclosed space

r is the volume rate at which air is recirculated

Equation 1 may be expressed as

$$\frac{dQ}{dt} = A - BQ \tag{1a}$$

where $A = ab\ (1 - E) + G$

$$B = \frac{Er + b}{V}$$

If a, E, G, B, b and r are treated as constants, disregarding transient variations in outdoor concentration or generation rate inside, or fluctuations in other parameters, equation 1a may be integrated

$$\int_{Q}^{Q_o} \frac{dQ}{A - BQ} = \int_{t}^{o} dt \quad , \tag{2}$$

or

$$\ln \frac{A - BQ}{A - BQ_o} = -Bt \tag{3}$$

where Q_o represents the amount of pollutant present in the enclosed volume at time $t = o$. Solving for

$$Q = \frac{A}{B}\left(1 - e^{-Bt}\right) - Q_o e^{-Bt} \tag{4}$$

If $t = \infty$, and a steady state has been reached,

$$Q_\infty = \frac{A}{B} \tag{5}$$

where Q_∞ is the value of Q when $t = \infty$. Although in theory infinite time is required for steady state to be reached, Q may closely approach Q_∞ in a comparatively short time.

b. Response Time

The response time of an enclosed space, for the purposes of this analysis, is defined as the time required for the inside concentration to go from zero to a given fraction of its steady state level. For example, $t_{.99}$ would be the time required for the inside concentration to go from zero to 99 percent of its steady state level. From equations 4 and 5,

$$\frac{Q}{Q_\infty} = 1 - e^{-Bt} + \frac{Q_o}{Q_\infty} e^{-Bt} \ , \tag{6}$$

which reduces to

$$e^{-Bt} = \frac{Q - Q_\infty}{Q_o - Q_\infty} \tag{6a}$$

and

$$t = \frac{1}{B} \ln \frac{Q_o - Q_\infty}{Q - Q_\infty} \tag{7}$$

For example, if $Q_o = 0$ and $Q = 0.99\ Q_\infty$

$$t_{.99} = \frac{1}{B} \ln \frac{0 - Q_\infty}{0.99\ Q_\infty - Q_\infty} = \frac{1}{B} \ln 100 \tag{7a}$$

or

$$t_{.99} = \frac{4.606}{B} \tag{7b}$$

It is to be noted that the response time is a characteristic of the building or enclosed space and its ventilation system. It is independent of the level of outside concentration. While the outside concentration is important in determining what the steady state indoor concentration will be, in this example, it does not determine how fast steady state will be reached.

c. Relationship Between Indoor and Outdoor Concentration

If equation 5 is expanded into its individual elements

$$Q_\infty = \frac{A}{B} = \frac{ab\ (1 - E) + G}{\frac{Er + b}{V}} \tag{8}$$

$$\frac{Q_\infty}{V} = \frac{ab\ (1 - E)}{Er + b} + \frac{G}{Er + b} \tag{9}$$

This reduces to the equation for a straight line between the inside concentration, $\frac{Q_\infty}{V}$, and the outside concentration, a.

$$\frac{Q_\infty}{V} = Ma + N \tag{9a}$$

where the slope,

$M = \frac{b\ (1 - E)}{Er + b}$, is the filtering factor, and

the intercept

$N = \frac{G}{Er + b}$, is the source-sink factor.

If the parameters in the foregoing model can be evaluated, it provides a basis for predicting the relationship between indoor and outdoor pollution, and for estimating how rapidly a building would approach equilibrium with its surroundings. It also provides a basis for estimating the effect of changing parameters such as filter efficiency or relative amounts of outside air and recirculated air on the

indoor pollutant level and on the response time of the building.

d. Application to Buildings and Individual Ventilating Systems

Large buildings with central air conditioning systems usually contain several air handling units, so that the parameters in the model would represent average values for all of the air handling systems and the spaces they serve. It also may be useful to apply the model to individual air handling units. In this case there is interchange between adjacent systems in the building which is not included in equation 1. If p is the rate of interchange between and air handling system and its neighbor, pollutant will be brought into the system at a rate Cp, where C is the concentration of pollutant in the adjacent system, and when there is more than one adjacent system there must be a separate term for each system. Pollutant will be lost to the adjacent system at a rate $\frac{Q_p}{V}$. If the rate of exchange is small, or if the concentrations in adjacent systems are nearly equal, the effect of this interchange will be small. On the other hand, if neither of these conditions is met, interchange between systems becomes an important consideration.

e. Application to a Fluctuating Outside Pollutant Level

The foregoing analysis is applied to the case where the outside pollutant level is constant. If the response time of the building is short compared with the time over which fluctuations occur, the assumption of constant outside concentration is realistic for purposes of this analysis. However, if the changes in outside concentration are not slow with respect to the response time of the building, further analysis of the intrusion of pollutant is required.

For descriptive purposes consider the case where the outside concentration fluctuates as a sine function. That is,

$$a = a_1 + a_o \sin \omega t \tag{10}$$

where a_1 is the average concentration, a_o is the amplitude, and ω is the angular rate of change of the sine function. Equation 1a is then modified to

$$\frac{dQ}{dt} = A' + A'_o \sin \omega t - BQ \, , \tag{11}$$

or

$$\frac{dQ}{dt} + BQ = A' + A'_o \sin \omega t, \tag{11a}$$

where

$$A' = a_1 \, b \, (1 - E) + G \, , \text{ and}$$

$$A'_o = a_o \, b \, (1 - E)$$

Solving for Q,

$$Q = \frac{A'}{B} (1 - e^{-Bt}) + Q_o e^{-Bt} + \frac{A'_o}{B^2 + \omega^2} \; e^{-Bt} + \frac{A'_o}{B^2 + \omega^2} (B \sin wt - w\cos wt) \tag{12}$$

Over an integral number of cycles

$$B \sin \omega t - w\cos \omega t = o$$

Also a cumulative average of the sin-cos term with time, which is applicable to the case where concentration is averaged over a long period of time, approaches zero asymptotically. Also $e^{-Bt} \rightarrow o$ with increasing time. And

$$Q_\infty = \frac{A'}{B} \tag{13}$$

which is similar to equation 5, except that average concentration a_1 is substituted for a.

If equation 12 is considered only at integral numbers of cycles, this is equivalent to considering only average values of Q and ignoring the periodic fluctuations. Under these conditions it can be shown that

$$t = \frac{1}{B} \ln \frac{Q_o' - Q_\infty}{Q - Q_\infty} \tag{14}$$

where $Q_o' = Q_o + \frac{A'}{B^2 + \omega^2}$. This is analogous to equation 7. It suggests that with a cyclic outdoor concentration the average amount of pollutant indoors reaches a given fraction of its steady state value in less time when the outdoor concentration is constant.

Actual outdoor pollutant levels usually do not fluctuate as simple sine functions, but the foregoing analysis suggests, at least descriptively, that if indoor and outdoor concentrations are averaged over a sufficient time, average values should approach a linear relationship analogous to equation 9.

f. Filter Efficiency

In the foregoing analysis filter efficiency is treated as a constant. However, it is possible in some cases that the first pass efficiency, when the pollutant concentration is highest, may be greater than the efficiency in subsequent passes through the filter. In the case of particulates this is true, because most of the lint and large particles are removed in the first pass, and what remains in suspension is mainly smaller particles. If the simplifying assumption is made that the filter has an average first pass efficiency of E_1, and an average efficiency of E_2 in subsequent passes, equation 1 becomes

$$\frac{dQ}{dt} = ab\,(1 - E_1) + G - \frac{QE_2r}{V} - \frac{Qb}{V} \tag{15}$$

and

$$A = ab\,(1 - E_1) + G$$

$$B = \frac{E_2 r + b}{V}$$

This is equivalent to assuming that the filter has a different efficiency towards outside air than towards return air. Since E_2 is usually less than E_1, B would also be smaller, and response time calculated from equation 7 would be longer. Furthermore equation 9 would become

$$\frac{Q_\infty}{V} = ab\,\frac{(1 - E_1)}{E_2 r + b} + \frac{G}{E_2 r + b}$$

In other words, a decrease in filter efficiency after the first pass would lead to a different value of some of the other parameters, but would not change the form of the equations.

g. Non-Airconditioned Buildings

The "black box" model is applied to buildings with forced ventilation, which includes most buildings with central airconditioning. In buildings without forced ventilation infiltration plays a dominant roll in determining exchange of air with the outside. While it is possible to model such a process, it is difficult to evaluate the parameters, because infiltration is subject to many uncontrolled variables. The understanding of the intrusion of pollutants into homes and other non-airconditioned buildings is of great practical importance, but it requires a better quantification of the infiltration process, and this is part of another study which has been initiated in this section. In any case Q in equation 1 is taken as the total amount of pollutant in the enclosed space, consisting of gaseous and particulate components, regardless of the manner the pollutants entered the space.

C. Indoor/Outdoor Dust Measurements

Measurements have been made of dust levels in the return air and the supply air of the Building Research Building with paper tape samplers and high volume samplers. In these comparisons the outdoor concentrations were higher than the indoor concentrations. The differences were much greater using the high volume samplers which are based upon weight and assign greater importance to large particles than by paper tape samplers which are based on optical density of the collected spots.

Recent work presented at the January 1971 meeting of ASHRAE also reported higher average dust levels outdoors than indoors (5, 6, 7). On the other hand earlier work by Jacobs et al (8) reported several comparisons in which indoor concentrations were higher, although average of all their comparisons indicated slightly less dust indoors than outdoors.

As a test of equation 9a indoor dust levels were plotted against outdoor levels using the data collected at the Building Research Building. The data showed considerable scatter. This suggests that more attention will have to be given to the effect of variations in the level of indoor activity.

D. Indoor Particle Generators

Another type of particle measurement has been made in which a light scattering particle counter has been placed in an unventilated room, and the rise and fall of dust count made before, during, and after particle generating operations such as the smoking of a cigarette, vacuum cleaning a rug, frying grease in a pan, or operating an oscillating electric fan. These are all common indoor operations, and the results indicate that smoking, and frying generate more of a fine particle population which remains in suspension for a long time than vacuum cleaning or operating an electric fan. In fact the increase in particle count produced by operating a fan begins to fall off and returns to initial levels even with the fan still operating.

E. Experimental Flow Measurements

Two of the important parameters in predicting the performance of a building or ventilating system are b, the rate at which fresh air is drawn into a building, and r, the recirculation rate. Work is in progress to develop techniques for measuring these quantities.

One suggested approach is the measurement of the air temperature of the fresh air, return air, and supply air streams. In other words, if one part of outside air and X parts of return air come into the air handling unit,

$$t_o + Xt_t = (1 + X)\,T$$

and

$$X = \frac{T - t_o}{t_r - T}$$

where t_o is the temperature of the outside air, t_r is the temperature of the return air, and T is the temperature of the total supply air.

For example, if outside air temperature is 30 °F, return air temperature is 70 °F, and the ratio return air to outside air flow is 4, T = 62°. $t_r - T = 70 - 62 = 8$ °F. Experimental measurements of T in the chamber downstream from the filters gave a range of values of greater than 10 °F. Therefore poor air mixing places a serious limitation on this method for measuring relative airflow rates.

Work is in progress to explore anemometer scanning techniques for measuring the fresh air and return air flow rates. This is not a simple problem, because it is difficult to find places free of turbulence and irregular flow patterns. Experiments are in progress with lightweight portable ducts to reduce the problem of measuring flow from grills to a problem of measuring flow in a parallel duct.

F. Tracer Measurements

An electron capture detector capable of detecting subparts per billion levels of sulfur hexafluoride has been obtained. It is currently being tested against experimental concentrations of sulfur hexafluoride in the test duct. It is hoped that this device will prove useful in tracer studies and may also be useful for detecting other electron capturing species besides sulfur hexafluoride.

G. Analysis of Particulates

Infrared absorption measurements were made of dust samples collected from the return air and fresh air ducts in the Building Research Building with high volume samplers. Ammonium sulfate has been identified as one of the constituents of indoor dust by this technique. This has been presented in a prepared discussion of a paper by Yocum and Cote (7a) on indoor/outdoor air pollution which has been submitted for publication in the ASHRAE Transactions.

A powder x-ray diffraction pattern of these samples, performed in the Crystallography Section, indicated that quartz was an important constituent of outdoor dust. The indoor dust did not produce sufficiently defined peaks for identification by this method.

Infrared absorption spectra and x-ray diffraction are sometimes useful in identifying constituents of dust, but they do not identify all of the major constituents. Improvement in dust collection techniques might improve the usefulness of these methods for dust analysis to some degree. In the past more attention has been given to determination of carcinogens and deleterions components than to the determination of the principle constituents of dust. This may be one reason why it has been estimated that only about 35 percent of dust is accounted for in analysis (1a).

Emission spectra of these high volume samples and samples collected with paper tape samplers were performed in the Spectrochemical Analysis Section. The results indicated that lead was a consistent trace constituent of both indoor and outdoor dust. However, it was also evident from these trial measurements that there is room for improvement in the methods of collecting dust for analysis. The paper used in collecting paper tape samplers is not an absolute filter, while the glass fiber paper used in collecting high volume samples cannot be used for determining calcium, silicon, aluminum, and some of the lighter elements.

Emission spectroscopy is a sensitive method for identifying trace amounts of heavy metals. It finds its greatest usefulness when there is no prior knowledge of what elements are present. In analyzing for preselected elements other methods such as atomic absorption spectroscopy, ring oven techniques, and colorimetric methods may be useful.

H. Analysis of Gaseous Pollutants

SO_2 is the air pollutant which generally receives first attention, and it is one of the first pollutants for which clean air standards have been developed. However it is not always present in detectable amounts particularly during summer months (6). Spot checks of SO_2 at Gaithersburg, by the West Gaeke Method, indicated an outdoor concentration of 0.02 ppm in one measurement, while it was not detected indoors or outdoors in two other tests. While the sensitivity of the measuring technique could have been improved, SO_2 was not present in high concentrations at the Gaithersburg site.

A qualitative test in the Building Research Building by the Saltzman method indicated that NO_2 was present in detectable amounts.

There is no wet method for CO comparable in sensitivity with the West Gaeke method for SO_2 or the Saltzman method for oxides of nitrogen. However, accessory columns were prepared for determining CO by the gas chromatographic method of Porter and Volman (10) as modified by the Stevens et al (11). However, this work has not been pursued far enough to develop a measurement capability. Measurements with tubes similar to those described by Shepherd (12) indicated significant amounts of CO in a closed kitchen with a gas stove operating.

Preliminary attempts were made to use a gas chromatograph with a flame ionization detector as a total hydrocarbon analyzer. This was done by replacing the column by a glass capillary connected with teflon leads. Two closely spaced peaks were obtained suggesting some separation of components even with this simple non-selective column. The results suggest that there may be significant differences between indoor and outdoor air with respect to the kind of pollutants which are detectable with a flame ionization detector.

I. Phthalates in the Laboratory

In 1965 it was reported by Blumer that laboratory solvents when evaporated to dryness left residues having infrared absorption spectra closely resembling those of certain phthalates (13). He suggested that the phthalates came from the adhesives used in the air filters. In a way this is surprising since phthalate esters are characterized by low vapcr pressure.

Spectra closely resembling those of Blumer have been produced in this laboratory from residues obtained by evaporating laboratory solvents. However, so far it has not been possible to demonstrate independently that the phthalates are airborne, because the residues cannot be reproduced at will. Ordinarily phthalates are not considered to be health hazards, but the problem is receiving attention, because impurities in laboratory solvents is undesirable per se.

3. Plans for Future Work

One of the tasks which is planned in the near future is to monitor dust indoors and outdoors with a particle counter and supplement this data with paper tape and high volume sampler data. As it stands indoor air usually contains less particulates than outdoor air, but this may not necessarily be true with respect to particles smaller than 1 μm. Smoking, for example, generates mostly small particles.

Comparison of the fresh air and return air provides a useful comparison of indoor/outdoor pollutant relationships. However, special attention will also be given to comparison of return air and supply air as a test of pollutant sources or sinks within a building. Allowing for a time lag for air to move from the supply back into the return, the air composition at these two points in the system should be identical in the absence of any source or sink.

Attempts will be made to measure the flow rates of the return air and fresh air in the air handling units. If this is successful it is a step towards quantitatively testing the model of an air conditioned building, and determining building response time.

It is planned to calibrate the electron capture detector for measuring sulfur hexafluoride so that it can be used for infiltration measurements. One of the problems to which it will be applied is the measurement of the rate of air exchange between different air handling units in a building.

It is also planned to explore the possibility of using "absolute" filter media in paper tape samplers with a view to capturing particulates for trace analysis. Microsorban, a benzene soluble filter medium, will be tested as a possible medium for both paper tape samplers and high volume samplers.

The foregoing tasks can be performed with available equipment. There are other areas however where the project's measurement capability must be improved before any extensive tasks can be undertaken, particularly in the analysis of gaseous pollutants. For example, it would be desirable to develop a strong gas chromatographic capability. This not only involves acquisition of instrumentation but also the development of expertise in its operation and in the design of special purpose modifications. This method has great versatility (14), and with the proper selection of columns and detectors it can be adapted to the measurement of many different pollutants. It is envisioned that its primary usefulness would be in looking for compounds and groups of compounds where there is no prior knowledge of what pollutants are present, somewhat in the way in which Dravnieks and Whitfield (15) have applied it to the study of odors. It would also be a monitor for compounds for which commercial recording monitors of sufficient sensitivity and specificity are not available or where there is only occasional monitoring need.

As monitoring needs are identified it would also be desirable to acquire recording monitors such as those available for NO_2 and NO and the flame photometric monitor for SO_2, the detector of which might also double as a chromatographic column detector for other sulfur compounds. A Mast ozone analyzer is already available in this laboratory, and its usefulness would be enhanced by a compatable recorder. The Mast analyzer is an iodometric instrument and is not specific for ozone. However, it is capable of making useful comparisons and providing a check on possible ozone generation by equipment such as copy machines and electrostatic filters. SO_2, oxides of nitrogen, and ozone are mentioned at this point, because they are pollutants for which no adequate gas chromatographic methods have been available for measuring sub ppm concentrations. However, Stevens, et al have recently described a gas chromatographic technique for separating H_2S, SO_2, CH_3SH, and C_2H_5SH at ppb levels(27).

The study of indoor air pollution often requires concurrent sampling at more than one site. In the case of paper tape samplers and high volume samplers this is accomplished by using a separate sampler at each site. As monitors become more sophisticated and more expensive this adds to monitoring cost, and other procedures for multisite sampling must be considered. One possibility is to develop automated switching and valving networks leading from more than one site into a single monitor. This procedure is limited to pollutants which are not lost or decomposed in transit through the network. In some instances it may be possible to collect samples in plastic bags using sequential samplers as was done by Ajemian and White (17) in monitoring carbon monoxide. It is also desirable that monitoring equipment be as portable as possible and, if not hand carried, at least mobile. In this way it would also be possible to obtain concurrent multisite time series data from a number of sites by visiting them with a single monitor in a timed sequence.

For some purposes non-automated wet methods might be used, and this would require equipment such as impingers, pumps, and flow measurement and control devices.

While the need for improvement in gaseous pollutant measurement capability is greatest, particle measurement capability could also be improved. For example, the lower measurement limit of light scattering counters is usually 0.3 μm. It would be desirable to extend this range to smaller sizes by means of a condensation nucleii counter which can measure sizes at least an order of magnitude smaller. Particles smaller than 0.3 μm are retained in the lungs as well as slightly larger particles and are not simply reexpelled as was once supposed.

More could be done in the analysis and identification of dust. Microscopic identification along the lines suggested in the Particle Atlas (17) and acquisition of collections of identified particles would be helpful. However, many of the particles in natural dust are too small to be identified by light microscopy, and electron microscopy is required. One problem is to collect dust samples in a form

suitable for electron microscopic examination. Particle capturing techniques such as specially designed electrostatic precipitators (18) or thermal precipitators (19) have been used for the purpose.

Atomic absorption spectroscopy has been used in the analysis of airborne dust for selected heavy metals (20), and acquisition of this capability would enhance the versatility of the project. This requires both instrumentation and the development of expertise in the collection and preparation of samples for analysis. Also the use of the ring oven technique (4c) might be explored as a method of analysis.

Particle counters provide size distribution data but do not provide information as to the chemical composition of particles as a function of size. The use of particle classifying devices for collecting samples for analysis should be explored. The Andersen sampler (21) and the Lundgren impactor (22) have been used for this purpose.

The benzene soluble fraction of dust should be examined, because benzo (α) pyrene and other carcinogens have been identified in this fraction. Yocum and Cote (7) usually found a higher benzene soluble fraction in indoor air than in outdoor air. Also Sawicki (23) has mentioned that the concentration of carcinogens is greater in indoor dust. Thin layer chromatography, fluorometry, and gas chromatography have been important techniques in the measurement of these materials.

Air ions are not ordinarily regarded as pollutants, but they are biologically active and can reputedly make a difference in heart rate and other bodily functions. Albert P. Kreuger has been active in research in this field, and a popular account of air ions has been presented under the auspices of the University of California (24). It would be desirable to make indoor/outdoor comparisons of the relative number and sign of air ions. At present a Royco ion collector for measuring air ions and a Ziesse-Penney (25) apparatus for measuring net charge on dust are available in this laboratory, but they require a sensitive electrometer and suitable recorder for monitoring.

The latter part of this report is presented as an inventory of possible equipment and types of measurements which might be made to augment dust measurements and flow measurements that are in progress. In fact, acquisition of gas chromatographic and certain other monitoring capability is essential if any extensive survey of gaseous pollutants is undertaken. This inventory is not exhaustive as to the types of equipment or techniques which may be applied to air pollution measurement, but acquisition of any of these devices and development of the techniques for their operation would strengthen the indoor pollution measurement capability.

4. Acknowledgment

The authors are indebted to Dr. Tamami Kusuda, particularly for helpful assistance in the derivation of the indoor/outdoor pollution model.

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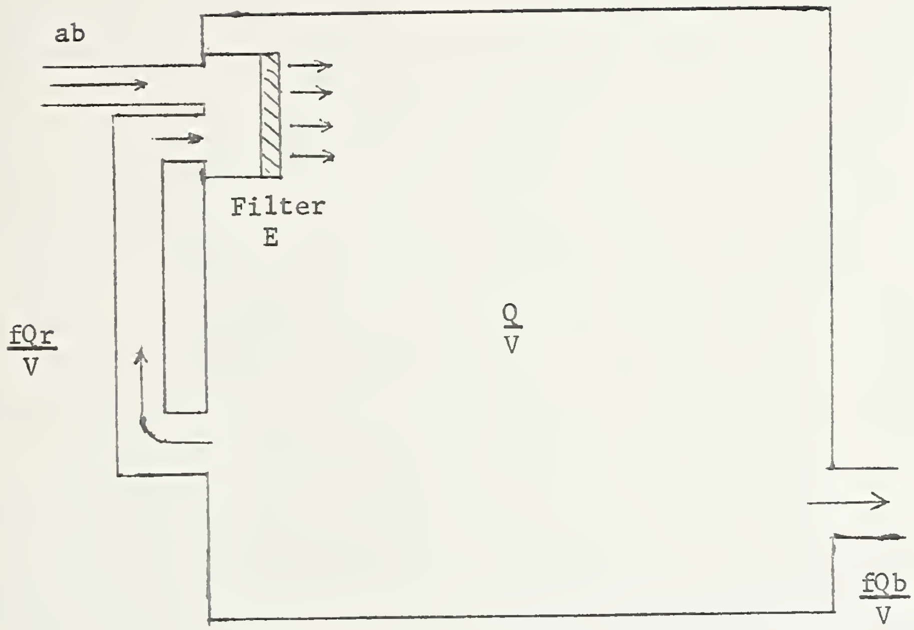

Figure 1 Diagrammatic Representation of an Enclosed Space with Forced Ventilation